Two-photon transitions in Ca^+, Sr^+, and Ba^+ ions

W. R. Johnson*
Department of Physics, University of Notre Dame, Notre Dame, IN 46556

U. I. Safronova
Physics Department, University of Nevada, Reno, NV 89557

M. S. Safronova
Department of Physics and Astronomy, 217 Sharp Lab, University of Delaware, Newark, Delaware 19716

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Two-photon 2E1 decay rates are calculated for the metastable $3d_j$ states in Ca^+, $4d_j$ states in Sr^+, and $5d_j$ states in Ba^+ to evaluate contributions of these transitions to the corresponding lifetimes. The calculations are carried out using the relativistic single-double method, where single and double excitations of Dirac-Fock wave functions are included to all orders of perturbation theory. We find that these rates are affected extremely strongly by the inclusion of the correlation corrections.

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I. INTRODUCTION

Recent advances in high-precision theoretical and experimental methodologies allowed for significant improvement in determining the lifetimes of long-lived metastable nd states in Ca^+, Sr^+, and Ba^+. These ions are of particular interest for developing optical frequency standards [1–3] and quantum information processing [4] owing to the extremely long lifetimes of nd states. High-precision calculations and measurements of these lifetimes are subject of a large number of publications. One of the first theoretical values of the nd state lifetimes in Ca^+, Sr^+, and Ba^+ was published by Guet and Johnson [5]. Theoretical and experimental studies of these lifetimes were presented in Refs. [6–22] for the $3d$ states of Ca^+, in Refs. [23–26] for the $4d$ states of Sr^+, and in Refs. [27–36] for the $5d$ states of Ba^+. Theoretical results for all three of these systems together with the review of previous theoretical calculations and available experimental measurements were presented by Sahoo *et al.* [37].

The most recent theoretical and experimental values of these lifetimes are summarized in Tables I and II. The experimental values of Ba^+ $5d$ lifetimes have the largest uncertainties (5-15%) since these lifetimes are by far longer than the corresponding lifetimes in the other two systems leading to complications in experimental measurements. From the theoretical standpoint, expected precision is similar in all three systems. The other four lifetimes were recently experimentally determined with better than 1% uncertainty. A high-precision result (0.4%) for the lifetime of the $4d_{5/2}$ level in Sr^+ was presented by Letchumanan *et al.* [26]. Uncertainties of 0.6% and 0.7% were quoted for the lifetime of the $3d_{5/2}$ level in Ca^+ by Barton *et al.* [18] and Kreuter *et al.* [22], respectively. The uncertainties of the theoretical $3d_{5/2}$ lifetimes in Ca^+ calculated by Kreuter *et al.* [22] and by Sahoo *et al.* [37] were estimated to be 0.9% and 0.8%, respectively. Theoretical and experimental values of the $3d_{3/2}$ and $3d_{5/2}$ lifetimes in Ca^+ from Ref. [22] are in agreement within the uncertainty bounds. However, theoretical coupled-cluster value of the $3d_{5/2}$ lifetime in Ca^+ given by Sahoo *et al.* [37] differs from the experimental value [22] by 4.3%. The difference between precision theoretical and experimental values of the $4d_{5/2}$ lifetime in Sr^+ presented by Sahoo *et al.* [37] and by Letchumanan *et al.* [26] is equal to 5.1%. The all-order theoretical Sr^+ lifetimes [38] are in agreement with experimental values.

We note that the theoretical uncertainty of the lifetimes values only include the estimated uncertainty of the primary $(n+1)s - nd$ E2 transitions. Some significant discrepancies in the lifetimes of the upper $5d_{5/2}$ level in Ba^+ were later explained by the contribution of the $5d_{5/2} - 5d_{3/2}$ M1 transitions [39]. Other transition possible M1 and E2 transition rates were evaluated and their contributions were found to be negligible [37]. In light of the improved precision of theory and experiment, as well as some remaining discrepancies, it is important to consider the possibility that other processes may contribute to the lifetimes of the nd metastable levels. It is particular important since the lifetimes are so long, in particulars in the case of $5d_{3/2}$ state in Ba^+ which lifetime is over 80 seconds.

In the present work, we consider the two-photon $(n+1)s - nd$ decays in Ca^+ (n =3), Sr^+ (n =4), and Ba^+ (n =5) ions via two E1 dipole transitions involving $n'p_j$ states. The singly ionized Ca, Sr, and Ba atomic systems are monovalent systems with a single valence electron outside of a closed core. The simplest one-electron system is atomic hydrogen. Two-photon transitions in H-like ions are among most widely studied atomic transitions, both theoretically and experimentally

*Electronic address: johnson@nd.edu; URL: www.nd.edu/~johnson

TABLE I: Lifetimes τ of the $nd_{3/2}$ states in Ca^+, Sr^+, and Ba^+ in seconds.

Ion	State	Theory	Experiment
Ca^+	$3d_{3/2}$	0.98 [6]	1.111±0.046 [11]
		1.271 [5]	1.17±0.05 [16]
		1.16 [8]	1.20±0.01 [18]
		1.080 [9]	1.176±0.011 [22]
		1.196±0.011[22]	
		1.185±0.007[37]	
Sr^+	$4d_{3/2}$	0.454 [5]	0.435±0.004[24]
		0.422 [25]	0.435±0.004[25]
		0.426±0.007[37]	0.455±0.029[25]
		0.441±0.003[38]	
Ba^+	$5d_{3/2}$	83.7 [5]	79.8±4.6 [31]
		81.5 [33]	89.4±15.6 [35]
		81.4 [34]	
		80.086±0.714[37]	
		82.0 [35]	
		81.5±1.2[36]	

TABLE II: Lifetimes τ of the $nd_{5/2}$ states in Ca^+, Sr^+, and Ba^+ in seconds.

Ion	State	Theory	Experiment
Ca^+	$3d_{5/2}$	0.95 [6]	0.994±0.038 [11]
		1.236 [5]	1.064±0.017 [14]
		1.14 [8]	0.969±0.021 [15]
		1.045 [9]	1.09±0.05 [16]
		1.165±0.011 [22]	1.100±0.018 [17]
		1.110±0.009 [37]	1.168±0.007 [18]
Sr^+	$4d_{5/2}$	0.405 [5]	0.372±0.025 [23]
		0.384 [25]	0.408±0.022 [25]
		0.357±0.012[37]	0.3908±0.0016 [26]
		0.394±0.003[38]	
Ba^+	$5d_{5/2}$	37.2 [5]	32±5 [28]
		30.3 [33]	34.5±3.5 [29]
		36.5 [34]	32.0±4.6 [35]
		29.856±0.296 [37]	
		31.6 [35]	
		30.4±0.4 [36]	

(see Refs. [40–54]). The 2E1 two-photon transition gives the dominant contribution to the lifetime of the $2s$ metastable state in H-like ions. With increasing nuclear charge Z, the importance of the one-photon magnetic-dipole (M1) transition increases as $\sim Z^4$ and it becomes dominant (70%) for Sn^{49+}.

To the best of our knowledge, no estimates of two-photon decay rates of the nd metastable levels of monovalent ions have been carried out prior to this work. In the present paper, we use expressions for the two-photon decay rates by performing summations over intermediate np_j states. The evaluation of the required reduced electric-dipole matrix elements is performed using the relativistic single-double (SD) all-order method, where single and double excitations of Dirac-Fock wave functions are included to all orders of perturbation theory. Final results for the two-photon 2E1 transition rates are calculated for the $3d_j$ states in Ca^+, $4d_j$ states in Sr^+, and $5d_j$ states in Ba^+.

II. METHOD

The 2E1 decay of excited state w to ground state v in an atom with one valence electron is given by the expression [55]

$$\frac{dW}{d\omega_1} = \frac{8}{9\pi}\alpha^6\omega_1^3\omega_2^3\sum_{q_1q_2}\left|M_{q_1q_2}\right|^2, \tag{1}$$

where the photon frequencies are related by energy conservation, $\omega_1+\omega_2 = E_f - E_i$. The two-photon matrix element $M_{q_1q_2}$ is given by

$$\begin{aligned} M_{q_1q_2} &= \sum_n \left[\frac{\langle w\left|D_{q2}\right|n\rangle\langle n\left|D_{q1}\right|v\rangle}{E_n+\omega_2-E_v}\right. \\ &+ \left.\frac{\langle w\left|D_{q1}\right|n\rangle\langle n\left|D_{q2}\right|v\rangle}{E_n+\omega_1-E_v}\right]. \end{aligned} \tag{2}$$

In this equation, n designates intermediate states, and D is the dipole operator. It is convenient to consider two terms in Eq. (2) separately, $M_{q_1q_2} = C_{q_1q_2} + E_{q_1q_2}$, when performing the angular reduction:

$$\begin{aligned} C_{q_1q_2} &= \sum_{nj} D^{nj}(\omega_2)\sum_{m_n}(-1)^{j_w-m_v}\begin{pmatrix} j_w & 1 & j_n \\ -m_w & q_2 & m_n\end{pmatrix} \\ &\times (-1)^{j_n-m_n}\begin{pmatrix} j_n & 1 & j_v \\ -m_n & q_1 & m_v\end{pmatrix} \end{aligned} \tag{3}$$

$$\begin{aligned} E_{q_1q_2} &= \sum_{nj} D^{nj}(\omega_1)\sum_{m_n}(-1)^{j_w-m_w}\begin{pmatrix} j_w & 1 & j_n \\ -m_w & q_1 & m_n\end{pmatrix} \\ &\times (-1)^{j_n-m_n}\begin{pmatrix} j_n & 1 & j_v \\ -m_n & q_2 & m_v\end{pmatrix}, \end{aligned} \tag{4}$$

where

$$D^{nj}(\omega) = \frac{\langle w\|\, D\,\|nj\rangle\langle nj\|\, D\,\|v\rangle}{E_{nj}+\omega-E_v} \tag{5}$$

and $\langle w\|\, D\,\|nj\rangle$, $\langle nj\|\, D\,\|v\rangle$ are the reduced electric-dipole (E1) matrix elements.

To evaluate $M = \sum_{q_1q_2}\left|M_{q_1q_2}\right|^2$ we perform the sums over q_1, q_2 and magnetic substates of v and w, and divide the result by $(2j_v+1)$. Performing the angular reduction yields:

$$\begin{aligned} M &= \frac{1}{2j_v+1}\sum_{m_vm_w}\sum_{q_1q_2}\left|M_{q_1q_2}\right|^2 = \frac{1}{2j_v+1} \\ &\times\sum_{nn'j}\frac{1}{2j+1}\Big(D^{nj}(\omega_2)D^{n'j}(\omega_2)+D^{nj}(\omega_1)D^{n'j}(\omega_1)\Big) \\ &+\frac{2}{2j_v+1}\sum_{nj}\sum_{n'j'}\begin{Bmatrix} j' & 1 & j_w \\ j & 1 & j_v\end{Bmatrix}(-1)^{j+j'}D^{nj}(\omega_2)D^{n'j'}(\omega_1). \end{aligned} \tag{6}$$

In the case of the $d_{5/2} - np_{3/2} - s_{1/2}$ 2E1 transition, we obtain

$$M = \frac{1}{2j_v+1} \sum_{m_v m_w} \sum_{q_1 q_2} |M_{q_1 q_2}|^2 = \frac{1}{24} \left[\sum_n D^{n3/2}(\omega_2) + \sum_n D^{n3/2}(\omega_1) \right]^2 . \quad (7)$$

The result for the $d_{3/2}-np_{3/2}-s_{1/2}$ and $d_{3/2}-np_{1/2}-s_{1/2}$ 2E1 transitions is more complicated:

$$M = \frac{1}{2j_v+1} \sum_{m_v m_w} \sum_{q_1 q_2} |M_{q_1 q_2}|^2 \quad (8)$$
$$= \sum_{nn'} \left[\frac{1}{16} \left(D^{n3/2}(\omega_2) D^{n'3/2}(\omega_2) + D^{n3/2}(\omega_1) D^{n'3/2}(\omega_1) \right) \right.$$
$$+ \frac{1}{8} \left(D^{n1/2}(\omega_2) D^{n'1/2}(\omega_2) + D^{n1/2}(\omega_1) D^{n'1/2}(\omega_1) \right)$$
$$+ \left(-\frac{1}{12} D^{n3/2}(\omega_2) D^{n'3/2}(\omega_1) + \frac{1}{6} D^{n1/2}(\omega_2) D^{n'1/2}(\omega_1) \right)$$
$$\left. + \frac{1}{12}\sqrt{\frac{5}{2}} \left(D^{n3/2}(\omega_2) D^{n'1/2}(\omega_1) + D^{n1/2}(\omega_2) D^{n'3/2}(\omega_1) \right) \right] .$$

Numerical evaluations of expressions in Eqs. (7) and Eq. (8) are similar to the evaluation of frequency-dependent polarizabilities in monovalent atomic systems (see, for example, Ref. [56]). The sums over n and n' in Eqs. (7-8) converge rapidly. Therefore, only a few terms need to be calculated accurately. The details of numerical evaluation of the two-photon transition rates are discussed in detail in the next section.

III. RESULTS AND DISCUSSIONS

In Table III, we list reduced electric-dipole transition matrix elements in Ca^+, Sr^+, and Ba^+ calculated using a relativistic SD all-order method (columns "SD"). Details of those calculations for the singly-ionized Ca, Sr, and Ba atomic systems were given in recent papers [36, 38, 57]. The $6s - np_j$ (n=6-9) electric-dipole matrix elements and $6s - nd_j$ (n=5-7) electric-quadrupole matrix elements in Ba^+ were calculated using the relativistic all-order method by Iskrenova-Tchoukova and Safronova [36]. The black-body radiation (BBR) shifts of the $5s-4d_{5/2}$ and $4s-3d_{5/2}$ clock transitions in $^{88}Sr^+$ and $^{43}Ca^+$ were calculated using the relativistic all-order method in Refs. [38, 57], respectively. The calculations of the BBR shifts involved the calculations of electric-dipole matrix elements needed for the present work.

In present paper, we extend those calculations to obtain all E1 matrix elements involved in the evaluations of two-photon transitions given by Eqs. (7-8). Additionally, we list the lowest-order (DF) reduced E1 matrix elements in Table III to illustrate the size of correlation corrections

TABLE III: Reduced electric-dipole transitions matrix elements (a.u.) in Ca^+, Sr^+, and Ba^+ calculated using relativistic SD all-order method (columns "SD"). The lowest-order DF data are given in columns "DF" to illustrate the size of correlation corrections.

np_j	$d_{j'} - np_j$		$np_j - s_{1/2}$	
	DF	SD	DF	SD
Ca^+	$3d_{5/2} - np_j - 4s_{1/2}$ transitions			
$4p_{3/2}$	-4.13479	-3.24523	-4.52694	-4.09886
$5p_{3/2}$	0.00109	0.17533	0.00805	-0.08894
$6p_{3/2}$	-0.04097	-0.09518	0.05016	0.11169
$7p_{3/2}$	0.03800	0.06466	-0.04583	-0.08981
$8p_{3/2}$	0.03205	0.04815	-0.03812	-0.07178
$9p_{3/2}$	-0.02689	-0.03785	0.03170	0.05859
Ca^+	$3d_{3/2} - np_j - 4s_{1/2}$ transitions			
$4p_{1/2}$	-3.08248	-2.41731	3.20119	2.89784
$5p_{1/2}$	-0.00626	0.12536	0.00613	0.07507
$6p_{1/2}$	0.02764	0.06827	0.04152	0.08516
$7p_{1/2}$	0.02657	0.04644	0.03631	0.06747
$8p_{1/2}$	0.02265	0.03461	0.02979	0.05361
$9p_{1/2}$	0.01911	0.02721	0.02460	0.04363
$4p_{3/2}$	-1.37635	-1.07884	-4.52694	-4.09886
$5p_{3/2}$	0.00080	0.05899	0.00805	-0.08894
$6p_{3/2}$	-0.01383	-0.03193	0.05016	0.11169
$7p_{3/2}$	0.01277	0.02166	-0.04583	-0.08981
$8p_{3/2}$	0.01075	0.01612	-0.03812	-0.07178
$9p_{3/2}$	-0.00902	-0.01267	0.03170	0.05859
Sr^+	$4d_{5/2} - np_j - 5s_{1/2}$ transitions			
$5p_{3/2}$	5.00253	4.14969	-4.92110	-4.35075
$6p_{3/2}$	-0.07576	-0.14195	0.16058	0.03406
$7p_{3/2}$	-0.08008	-0.07816	0.02782	-0.05261
$8p_{3/2}$	-0.06458	-0.05327	0.00477	-0.05346
$9p_{3/2}$	0.05195	0.03971	0.00145	0.04635
Sr^+	$4d_{3/2} - np_j - 5s_{1/2}$ transitions			
$5p_{1/2}$	-3.72922	-3.08300	3.48479	3.07837
$6p_{1/2}$	-0.02628	-0.07847	0.06642	-0.02476
$7p_{1/2}$	-0.04692	-0.04487	-0.00503	-0.06259
$8p_{1/2}$	0.04031	0.03099	0.01278	0.05428
$9p_{1/2}$	0.03316	0.02322	0.01276	0.04468
$5p_{3/2}$	-1.65717	-1.36941	-4.92110	-4.35075
$6p_{3/2}$	0.02843	0.05105	0.16058	0.03406
$7p_{3/2}$	0.02800	0.02758	0.02782	-0.05261
$8p_{3/2}$	0.02231	0.01864	0.00477	-0.05346
$9p_{3/2}$	-0.01786	-0.01383	0.00145	0.04635
Ba^+	$5d_{5/2} - np_j - 6s_{1/2}$ transitions			
$6p_{3/2}$	5.00115	4.11081	5.47757	4.70971
$7p_{3/2}$	0.54254	0.44891	0.26098	0.08682
$8p_{3/2}$	0.29760	0.22194	0.07861	-0.03310
$9p_{3/2}$	0.20348	0.14354	0.03795	-0.04379
Ba^+	$5d_{3/2} - np_j - 6s_{1/2}$ transitions			
$6p_{1/2}$	3.74545	3.05455	3.89092	3.33801
$7p_{1/2}$	0.35129	0.27697	0.06536	-0.06203
$8p_{1/2}$	0.19564	0.13345	0.00707	0.08753
$9p_{1/2}$	0.13457	0.08508	0.01441	0.07272
$6p_{3/2}$	1.63537	1.33402	5.47757	4.70971
$7p_{3/2}$	0.18636	0.15495	0.26098	0.08682
$8p_{3/2}$	0.10189	0.07653	0.07861	-0.03310
$9p_{3/2}$	0.06955	0.04940	0.03795	-0.04379

TABLE IV: Example of evaluation of the two terms $D^{n3/2}(\omega_1)$ and $D^{n3/2}(\omega_2)$ in Eq. (7) needed for the evaluation of 2E1 two-photon $5d_{5/2}-6s_{1/2}$ transitions rate in Ba^+. All matrix elements are calculated using SD all-order method, $E_n = E_{np_{3/2}} - E_{5d_{5/2}}$, $\epsilon = w_1 + w_2 = E_{5d_{5/2}} - E_{6s_{1/2}} = 5674.807$ cm^{-1} = 0.025856 a.u. (Ref. [58]). In this example, $\omega_1 = [1/100]\epsilon = 0.00025856$ a.u. and $\omega_2 = [99/100]\epsilon = 0.02327$ a.u. $D^{n3/2}(w) = \langle 5d_{5/2} \| D \| np_{3/2} \rangle \langle np_{3/2} \| D \| 6s_{1/2} \rangle / (E_n + w)$.

$np_{3/2}$	E_n [58]	$E_n + w_1$	$E_n + w_2$	$\langle 5d_{5/2} \| D \| np_{3/2} \rangle$	$\langle np_{3/2} \| D \| 6s_{1/2} \rangle$	$D^{n3/2}(w_1)$	$D^{n3/2}(w_2)$
$6p_{3/2}$	0.0742	0.0744	0.0998	4.111	4.710	260.2	194.1
$7p_{3/2}$	0.2020	0.2023	0.2276	0.449	0.087	0.193	0.171
$8p_{3/2}$	0.2550	0.2552	0.2806	0.222	-0.033	-0.029	-0.026
$9p_{3/2}$	0.2825	0.2828	0.3081	0.144	-0.044	-0.022	-0.020

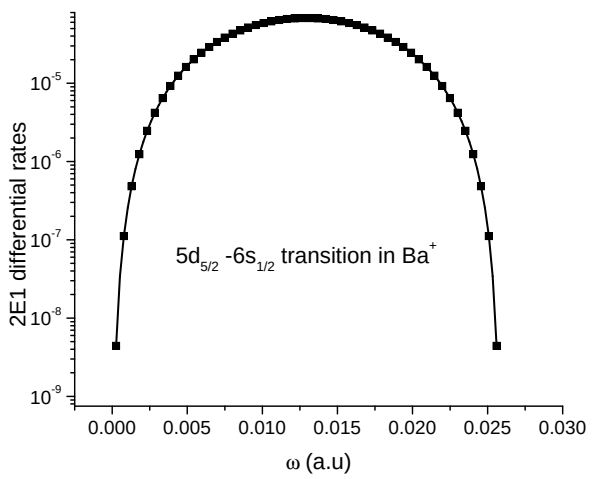

FIG. 1: Differential rate $\frac{dW}{d\omega}$ for two-photon decay of the $5d_{5/2}$ level in Ba II.

of individual matrix elements. The inclusion of correlation corrections significantly affect the two-photon transition rates.

In Table IV, we illustrate the evaluation of two terms $D^{n3/2}(\omega_1)$ and $D^{n3/2}(\omega_2)$ in Eq. (7) needed for the evaluation of 2E1 two-photon $5d_{5/2} - 6s_{1/2}$ decay rate in Ba^+. As discussed in the previous sections, only $5d_{5/2} - np_{3/2} - 6s_{1/2}$ decay channels are allowed in this case. All matrix elements listed in Table IV are calculated using SD all-order method, and experimental energies are used to evaluate $D^{n3/2}(\omega)$ terms. The value of $\epsilon = \omega_1 + \omega_2$ is equal to the energy difference between final and initial states. We use here the value ϵ = 5674.807 cm^{-1} = 0.02585632 a.u. from NIST website [58]. To perform numerical integration over ω_1 and ω_2 needed for the evaluation of total decay rate W is Eq. (1), we divide this energy difference ϵ into 100 intervals with the step $[1/100]\epsilon$ and calculate $D^{n3/2}(\omega_1)$ and $D^{n3/2}(\omega_2)$ at each point ($\omega_2 = \epsilon - \omega_1$). The values in Table IV are calculated for $\omega_1 = [1/100]\epsilon = 0.00025856$ a.u. and $\omega_2 = [99/100]\epsilon = 0.02327$ a.u. The sum over intermediate states converges extremely fast and nearly completely saturated by the first term $n = 6$. The quantities $\sum_n D^{n3/2}(w)$ only weakly depend on ω as the difference between $\sum_n D^{n3/2}(w)$ at the first ω grid point and last grid point is only 25%. The reason for such weak dependence is comparatively small interval $w_1 + w_2 = 0.0259$ a.u in comparison with the $E_{6p_{3/2}} - E_{5d_{5/2}}$ =0.0742 a.u. energy difference.

The final results for quantity M defined by Eg. (7) for the 2E1 $5d_{5/2} - 6s_{1/2}$ transition vary only weakly with ω (from 8607 to 8247 a.u.). Multiplying these values by a factor of $\frac{8}{9\pi}\alpha^6\omega_1^3\omega_2^3$ (see Eq. (1)) and integrating over w, we find the two-photon decay rate for the $5d_{5/2} - 6s_{1/2}$ 2E1 transition. In Fig. 1, we illustrate the differential rate $dW/d\omega$ for the 2E1 two-photon $5d_{5/2} - 6s$ transition in Ba II. Total 2E1 decay rate is equal to 8.079[-07] s^{-1}.

In Table V, we list the results for two-photon transition rates (s^{-1}) for the $4s - 3d_j$ transitions in Ca^+, $5s - 4d_j$ transitions in Sr^+, and $6s - 5d_j$ transitions in Ba^+. We note that the Sr^+ result was previously quoted in Ref. [38]. We list both the lowest-order DF results (in column labeled "Lowest-order") and our final SD all-order results (in column labeled "All-order"). We find very large difference, by a factor of 10-50, between the lowest-order and final all-order results. These differences are due in part to use of different $\epsilon = w_1 + w_2$ intervals in these calculations. These intervals are defined by the energy difference between the final and initial transition states, $E_{nd_j} - E_{(n+1)s}$. In the lowest-order calculation, the lowest-order values are used, and in the final all-order calculation, the experimental values of these intervals are used. For the example given in Table IV, the DF value of $\epsilon = 0.0350$ a.u is 1.35 larger than the experimental value. The values of reduced electric-dipole transition matrix elements also decrease with inclusion of the correlation effects (compare results in columns with 'DF' and 'SD' labels in Table III). As a result, the values of $\sum D^{n3/2}(w_1)$ and $\sum D^{n3/2}(w_2)$ decrease by a factor of two and the coefficient of $\frac{8}{9\pi}\alpha^6\omega_1^3\omega_2^3$ in Eq. (1) decreases by a factor of 5.6 in the case of the $6s - 5d_{5/2}$ transition in Ba^+.

IV. CONCLUSION

We have calculated two-photon decay rates for the $4s - 3d_j$ transitions in Ca^+, $5s - 4d_j$ transitions in Sr^+,

TABLE V: Two-photon decay rates (s^{-1}) for the $nd_j-(n+1)s$ transitions in Ba^+ ($n = 5$), Sr^+ ($n = 4$), and Ca^+ ($n = 3$) ions. The contributions of the 2E1 decay channel to the nd lifetimes are given in column labeled "Contr." in %.

Ion	Transition	Lowest-order	All-order	Contr.
Ca^+	$3d_{3/2} - 4s$	6.809[-3]	1.989[-4]	0.02%
Ca^+	$3d_{5/2} - 4s$	6.925[-3]	1.960[-4]	0.02%
Sr^+	$4d_{3/2} - 5s$	5.436[-3]	7.614[-4]	0.03%
Sr^+	$4d_{3/2} - 5s$	5.554[-3]	7.050[-4]	0.03%
Ba^+	$5d_{3/2} - 6s$	2.026[-5]	8.079[-7]	0.006%
Ba^+	$5d_{3/2} - 6s$	1.478[-5]	3.077[-7]	0.001%

and $6s - 5d_j$ transitions in Ba^+. We find that these rates are affected extremely strongly by the inclusion of the correlation corrections. The lowest-order calculation overestimates the values of these decay rates by 10 to 50 times. Our final all-order results snow that the contributions of the 2E1 decay channel to the lifetimes of metastable nd levels of Ca^+, Sr^+, and Ba^+ is negligible (0.001%-0.03%) at the present level of theoretical and experimental precision.

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